REVIEW

on **the competition for occupying the academic position professor** in the professional field 4.2. "Chemical sciences", scientific research area "Physical Chemistry" for the needs of the Institute of Physical Chemistry at the Bulgarian Academy of Sciences, Laboratory "Electron microscopy and microanalysis" - IPC. BAS on "Electron microscopy and simulation studies of phase formation processes in condensed matter".

announced in the State Gazette 20/10.03.2020

Candidate: **assoc. prof. Bogdan Stavrev Ranguelov, PhD**

Reviewer: **prof. Zara Petkova Cherkezova-Zheleva**, PhD, Institute of Catalysis - Bulgarian Academy of Sciences, member of the Scientific Jury

1. Background and brief biographical data about the applicant:

According to the Order № 57-РД-09/25.06.2020, issued by the Director of the Institute of Physical Chemistry, BAS (IPC-BAS), I was appointed as a member of the Scientific Jury for accomplishing the procedure in the competition for occupation of the academic position „professor" in the Institute of Physical Chemistry at the Bulgarian Academy of Sciences, in the professional field 4.2. "Chemical sciences", scientific research area "Physical Chemistry" for the needs of the Laboratory "Electron microscopy and microanalysis" - IPC. BAS on "Electron microscopy and simulation studies of phase formation processes in condensed matter", announced in the State Gazette 20/10.03.2020. Assoc. Prof. PhD Bogdan Stavrev Ranguelov is the only candidate, applying for the academic position "Professor" in the competition, announced by the IPC-BAS for the needs of the Laboratory "Electron microscopy and microanalysis".

Assoc. Prof. PhD Bogdan Ranguelov graduated Sofia High School of Mathematics in 1988 in a special class in physics. He graduated in physics from the Faculty of Physics of Sofia University "St. Cl. Ohridski" in 1995. In 2009 he was awarded with a PhD degree on the topic "Instability of vicinal crystal surfaces – step bunching" at the Institute of Physical Chemistry-BAS, in scientific research area "Physical Chemistry" with a diploma №: 33602 /07.12.2009 by the Higher Attestation Commission. After he won a competition in 2011 Dr. Bogdan Ranguelov became Associate Professor at IPC-BAS and currently he occupied this position as a head of the Laboratory of Electron Microscopy and Microanalysis. As of 19.05.2020 he has occupied the academic position Associated Professor for more than five years, which is the period required by the normative documents for the competition for the occupation of the academic position of "professor" at IPC-BAS. The applicant participated in research courses in well-known scientific centres abroad, which makes a significant contribution to his scientific development:

- Investigation of surface molecular reactions and catalytic properties of crystalline surfaces of Pt (111) and Fe (100) using HREELS (High resolution electron energy loss spectroscopy) and high vacuum technique - Friedrich-Alexander-Universität Erlangen-Nürnberg, Germany (2000-2004).
- Investigation of spiral growth on a Si (111) crystal surface, step-density waves using REM (reflection electron microscopy) and a directed diffusion of atomic clusters under the action of an external force using LEEM (Low energy electron microscopy) - Center Interdisciplinary of the Nanoscience of Marseille CINaM, Marseille, France (3 months, 2005 and 2018).

- Investigation of the processes of epitaxial growth of layers on the Si(111) crystal surface using REM - Institute of Semiconductor Physics, Siberian Branch of the Russian Academy of Sciences, Novosibirsk, Russia (2 weeks, 2010).

2. Review of the submitted documents:

Dr. Ranguelov has submitted a full set of application documents according to the requirements of the Regulation for the Terms and Procedure for Acquisition of Academic Degrees and for Occupation of Academic Positions in the Institute of Physical Chemistry of BAS. Assoc. Prof. PhD Bogdan Ranguelov meets the relevant law requirements for occupation of the academic position "Professor". He has submitted 41 scientific research publications in total. 23 of them were published after receiving of PhD degree and Assoc. Prof. academic position. They are an object of the current review. 8 of these publications are selected as an equivalent to a habilitation thesis, and another 15 original scientific papers cover the requirements of the present competition.

Based on the relevant summary submitted by the applicant it is obvious that he meets and even exceeds both the minimal National requirements and the specific requirements of IPC. BAS. It should be noted that his score for group Д, В, Г and E indicators is far above the relevant requirements. **The total score of the applicant is 923 points while the minimal required points are only 640.**

The total number of publications of Dr. Bogdan Ranguelov, submitted for participation in the competition is 41 and all of them are published in the editions refereed by Scopus / Web of Science. There is no evidence of plagiarism in all presented scientific works. The candidate applies in the competition with 23 papers in scientific journals refereed by Scopus / Web of Science database and 9 of them belong to the highest (Q1) category.

The submitted publications are divided into two groups, covering the В and Г indicators, according to the Rules of the conditions and the order for acquiring academic degrees and for taking up academic positions at the IPC - BAS. In the first group, indicator В–4 "Habilitation work - scientific publications in journals that are referred and indexed in world-famous scientific information databases (WoS or Scopus)" are presented 8 publications. They are valued at 160 points. According to this indicator, the candidate's score significantly exceeded the required 100 points. It should be noted that two of the 8 publications are in the Q1 area (WoS or Scopus), 4 are in Q2 area and two papers are in Q3 area, respectively. It should be also underlined that the half of these publications are developed in a team with only one co-author, and in 5 out of 8 the candidate is the first author, which clearly indicates his leading contribution to the preparation of the presented scientific papers. Dr. Ranguelov has made numerous studies in collaboration with leading international researchers. Fifteen scientific papers are presented in the second group covering Г-7 indicator. All of these papers are published in the referred journals in Q1 area (7 publications), in Q2 area (3 publications), in Q3 area (3 publications), in Q4 area (1 publication) and 1 publication with SJR only. So, the indicator Г has a total of 302 points, 220 points have been required. The advanced journals in the field of Q1, in which a large part of the results of Dr. Ranguelov's research activity have been published, are: Physical Review Letters, Surface Science, Ceramics International, ChemSusChem, Materials Letters, Journal of Non-Crystalline Solids, Journal of Environmental Chemical Engineering, Journal of the European Ceramic Society, International Journal of Hydrogen Energy, etc.

3. Analysis of the scientific and innovative research activities of the candidate and their applications in practice

The research and innovative activities of Dr. Bogdan Ranguelov are focused on the current topics of European and national priorities and they are in line with the recent scientific challenges. The investigations are related to scientific and applied electron microscopy studies, as well as to simulations of the phase formation processes in condensed matter. The

scientific interests of the candidate and his experience in the application of experimental and theoretical methods are focused on the study of the elementary mechanism of crystal growth, the dynamics of the atomic steps formation on the crystal surface, their spiral growth, etc. Dr. Ranguelov follows the traditions of one of the oldest and world-wide recognised Bulgarian School of nucleation and crystal growth established by I. Stranski and R. Kaishev. The fundamental and applied studies, presented by the candidate are also related to electron microscopy investigations of electrochemically prepared thin layers and electrodes, carbon nanomaterials, new and advanced synthered materials obtained from industrial and household waste in accordance with the green and the sustainable principles of circular economy, etc.

Research and applied research activities of the applicant are presented as oral and poster reports at more than 40 international and national scientific events. Assoc. Prof. Ranguelov selected a list of 10 oral presentations, which are given by him personally at key international and national scientific forums. Participation at prestigious scientific events and presentation of scientific papers as oral or poster reports has a significant impact on his scientific career, even that they have no formal reflection on the competition evaluation indicators. Dr. B. Ranguelov was recognizes as a leading scientist and he was elected as a Bulgarian representative and an executive committee member of the European Network of Crystal Growth.

Research and innovative activities of the applicant also include project activities. Dr. Ranguelov is a leader of one international research project DNTS/01/15/France and a team member of 11 international and national scientific or educational projects, as well as two EBR projects. The above mentioned scientific investigations and research papers are closely related to objectives and deliverables of the respective projects.

Educational and project activities of Dr. B. Ranguelov gave him a total of 185 points within the indicator E, while only 100 points are required. Dr. Ranguelov is a supervisor of one successful PhD student. In addition, the candidate is a participant in projects focused on young scientists training. He is also an organizer and lecturer of scientific schools with a main focus of PhD students and young scientists training. The candidate has not attached information about the financial earnings (within indicator E-18) of the Laboratory of Electron Microscopy and Microanalysis in IPC BAS, which is successfully managed by him up to this date. In this regard the real estimation in indicator E would have repeatedly exceeded the required 150 points.

Based on the achieved research and innovative results, Dr. Ranguelov has been registered in the Nacional Center for Information and Documentation - NACID (https://ras.nacid.bg/dissertation-preview/28364), where his doctoral degree and academic rank "associate professor" are recognized.

4. Evaluation of the scientific and innovative contributions of the candidate

The main scientific contributions of fundamental and innovative scientific research of Dr. Ranguelov can be summarized in the following **main thematic areas**:

1. Electron microscopy studies of phase formation processes in condensed matter (publications № 1-4, 11, 13-17, 20-22, 25-27, 30-34, 36-37, 40-41)

2. Simulation and theoretical studies of phase formation processes in condensed matter (publications № 5-10, 12, 18-19, 23-24, 28-29, 35, 38-39)

The main topic and the contributions of Dr. Ranguelov enrich and develop his scientific and applied contributions reached when he was awarded with PhD degree and academic rank of Associate Professor. The main research activities of the candidate are focused on the elementary processes that take place during the growth of vicinal crystal surfaces. Such a study is of particular importance, as each real crystal surface is practically a vicinal one and many steps with monoatomic height are formed on it. These monoatomic steps are moving during the crystal growth, which determines the morphology change. The

scientific contributions of the applicant in this area are undisputable. They increase the knowledge of the role of various factors in the complex processes of formation of monoatomic steps on the crystal surface. Factors that stabilize the micromorphology, migration of adatoms, their diffusion and incorporation into the steps, as well as the effect of "transparency" of the steps, etc. have been studied. It is important to emphasize that in these studies Assoc. Prof. Ranguelov combines two different research approaches. These are the theoretical simulation study and in-depth mathematical analysis of the monoatomic steps on the crystal surface. On the other hand, the obtained theoretical data is supported by a carefully designed experiment with modern equipment to illustrate the dynamics of the studied processes.

The **first main thematic area: electron microscopic studies of phase formation processes in condensed matter**, includes electron microscopic studies of two-dimensional nucleation processes, crystal growth and instability processes at vicinal crystal surface of Si (111) (publications № 4, 16, 20, 27, 29). Advanced equipment of *in situ* reflection electron microscopy (REM) was used and important experimental results were obtained for two-dimensional nucleation, for multilayer island and spiral crystal growth, as well as for critical width of terraces at vicinal crystal surface Si (111). New technique of low distortion reflection electron microscopy (LODREM) was used and undistorted images of grown monatomic spirals (with a step height of one lattice parameter) at a vicinal Si(111) crystal surface were obtained for the first time. Varying a temperature in a wide range and using the maximum achievable values for saturation, the processes of crystal growth are studied by a spiral mechanism. The density of the dislocation sources was estimated, as well as the number of left and right rotating spirals. The shape of the spirals was determined as Archimedean one. The overlap of the diffusion fields of adjacent steps has been confirmed due to the large mean free path of the adatoms in the respective high-temperature interval, the presence of the so-called back-stress effect, as well as the effect of "transparency" of the steps of the vicinal crystal surface with respect to the adatoms. The relationship between theoretical (simulation) studies [16, 20, 27] and experimental studies performed using REM [6, 9] shows details of homoepitaxy on vicinal Si(111) surface. Investigations [10, 29] found an experimental evidence of theoretically (by simulations) "predicted" new type of instability during crystal growth, the so-called "step-density waves ". The main subject of investigation is the so-called "critical terrace width". The cases of diffusion and attachment-detachment limited regimes of growth at high and low temperatures are registered. Parameters that allow the calculation of the critical nucleus size and the activation energy for two-dimensional nucleation have been also obtained. The morphology of the growing two-dimensional islands was studied by atomic force microscopy also. Similarly the so-called "transparency" of the steps was studied and in this regard wide terraces (about 100 micrometers) were prepared, which is a complicated experimental task [27].

Experimental observation and registration of step-density waves is a serious scientific challenge that the candidate has successfully tackled. It is related to finding of conditions in which there are no destabilizing factors and at the same time the REM equipment is used to monitor the growth dynamics of monatomic steps. The REM equipment was used for investigation of the "transparency" of the steps with a respect to the obtained pyramidal structures and the dependence of their morphology on the balance of the adatom flows [27].

An important part of the scientific and applied contributions of the candidate is related to the treatment of household waste and its usage as a raw material for the production of sintered glass-ceramic materials. Important technological results related to environmental protection and circular economy are obtained in papers [14, 15, 17, 25, 30, 32, 36, 40]. It was also studied the preparation of glass-ceramics by utilisation of accumulated large amounts of metallurgical waste (mainly iron oxides) during the production of steel and ferronickel [30,

36]. Another study investigates the conductivity and magnetic properties of materials obtained by the so-called Laser Floating Zone method [40].

The candidate has also a scientific contribution to the investigation of soft condensed matter [22, 37]. Two-antennary oligoglycines have been adsorbed on number of substrates using different concentrations. The obtained pool of experimental data shows the formation of a wide range of structural reorganiations. This opens interesting options in view of a number of innovative applications in medicine, pharmacy, tissue engineering, catalysis and biocatalysis, removal of various inorganic and biological impurities from aqueous media. The antibacterial effect against *E. coli* of ion-exchanged clinoptilolite (with an addition of Ag and Cd) was studied. Assoc. Prof. Ranguelov is undoubtedly an expert in the field of electron microscopy. He also participates in research teams for the investigation of structure and morphology of thin films and catalysts [1, 2, 3, 11, 13, 21, 26, 31, 33, 34, 41].

The candidate has significant scientific contributions in the **second studied thematic area: Simulation and theoretical studies of phase formation processes in condensed matter.** Theoretical approach (using simulations) was applied to investigate the instability of crystal growth on a vicinal crystal surface due to the appearance of a step-density wave and experimental evidence for this has been obtained [7, 8, 10, 29]. Modelling of the ability of the step to "accept" ("donate") adatoms during the crystal growth process is important for determining the morphology of crystal surface. In paper [7], the step dynamics during crystal sublimation and growth was studied in the limit of fast surface diffusion and slow kinetics of atom attachment-detachment at the steps. This model was developed in [8] by adding the so-called electromigration force. A transient growth model is developed in [10] for the limit of slow kinetics at the steps and fast surface diffusion. This model predicts instability of the vicinal surface but rather different expression for the wavelength of the most unstable mode, which depends only on the ratio between the step transparency and the step kinetic coefficients. Taking into account the influence of the transparency of the steps, the speed of a step at a time of day depends on the time (jumps), they are listened to at "more distant steps" in previous moments. The so-called "non-local" dynamics of the steps is studied by the Monte Carlo method [9]. Some conclusions concerning the stability of the vicinals are drawn. The picture of step bunching phenomena was additionally studied in [5, 6, 12, 18]. The maximum influence of the transparency of the steps was estimated in the temperature interval between kinetic and diffusion regimes [20]). The diffusion of adatoms and atom clusters at vicinal crystal surface, as well as the related phenomena, such as step transparency and change of cluster shape due to electromigration force, were studied by using Monte Carlo simulations [9, 23, 24, 38]. It was shown that even a very small value of electromigration force has a significant effect on the trajectory of adatom's diffusion. Papers [23] and [24] are focused on the diffusion of adatoms at a model terrace with (111) orientation by using the so-called tight-binding potential and Metropolis algorithm - Monte Carlo method. Important results on the shape changes of two-dimensional atomic islands and vacancy clusters diffusing on epitaxial (111) interfaces under the impact of an external force were obtained in [38] using Kinetic Monte Carlo simulations.

Monte Carlo simulations of the thermal stability and spontaneous breakdown of free-standing monoatomic metal nanowires [28] and of two-dimensional nanowires on homoepitaxial interfaces on fcc (111) interface [35] were performed, using Monte Carlo sampling, with many-body tight-binding second-moment approximation potential between the interacting atoms. A novelty in the paper [19] is modification of the classical diffusion limited aggregation (DLA) model in the attachment to the cluster rules and in the scheme of particle generation/killing. Thus the model is capable of producing a great variety of growing pattems (fractals, spirals) by changing only a single parameter and a morphological diagram of generalized DLA model with two different types of particles can be constructed. Publication

[39] includes the latest simulation studies of the candidate examining soft condensed matter and the interactions between model molecular complexes, the so-called "model of patchy particles". This paper attempts to elucidate the polymorph selection and crystal growth process in proteins and colloidal crystals using 2D Monte Carlo simulations and a computational model with short-range attraction for "protein-like' patchy particles of a specific patch geometry, bond width and strength.

All mentioned original scientific and applied contributions of Dr. Ranguelov open opportunities for a future research and an improvement of the fundamental knowledge about the elementary processes of single crystal growth with important technological applications. The research activities of the applicant include also contributions of significant practical importance in the field of growth of silicon single crystals, which is the basis of many modern technologies for the semiconductor device industry, solar cells manufacturing, etc. The design of single-crystal morphology is an important part of the miniaturisation of computers and high-tech devices together with the increasing of their high-performance. The theoretical analyzes in papers are mainly made for silicon surfaces, but they concern the crystal growth process in general.

5. International and local recognition and significance of the candidate's scientific results

The evidence of the international recognition and significance of the scientific results of Dr. Ranguelov's research works is the large number of citations. The total number of noticed citations of 24 publications with his participation at the date of submitting the documents is 198 according to databases Scopus/Web of Science. 18 papers included in the competition, were mentioned at 113 citations in Scopus/Web of Science databases. Thus, under indicator Д the candidate receives 226 points, which exceed the required 120 points for the position of "professor" in accordance with the Rules for the conditions and the order for acquiring scientific degrees and for occupying academic positions at IPC – BAS.

6. Critical remarks and recommendations to the scientific works of the candidate:

Dr. Bogdan Ranguelov is an established scientist. His theoretical and experimental research investigations are planned precisely and the obtained results are in-depth analyzed and interpreted adequately. The presented list of candidate's scientific contributions reflects accurately the achievements and marks new challenges in related scientific problems. I have no critical remarks and recommendations for the candidate's scientific work.

7. Personal impressions of the reviewer about the candidate:

Based on the documents submitted in the competition and my personal impressions, I recognize Assoc. Prof. Dr. Bogdan Ranguelov as a leading researcher with a clear scientific focus, whose competence I highly value. The results of his research have significant and undisputable scientific and applied contribution. They are mainly related to elucidation of the processes of phase formation in condensed matter through theoretical calculations and simulations, as well as by planning and conducting high-tech scientific or applied electron microscopy studies. The scientific works of the candidate, presented for participation in the competition, are entirely in the field of the thematic area of 4.2. Chemical sciences and research area "Physical Chemistry". The topics of his research fully correspond to the topics of the Laboratory of Electron Microscopy and Microanalysis, for which the competition was announced. The proficiency of Assoc. Prof. Ranguelov and his international recognition will undoubtedly increase the scientific level of the laboratory.

In conclusion, based on the review of the submitted documents for the announced competition and my personal opinion, it can be concluded that the score of Dr. Bogdan Ranguelov significantly exceeds the minimum required points for the occupation of the

position of "professor" in accordance with the Rules for Acquisition of Academic Degrees and for Occupation of Academic Positions in the IPC-BAS.

CONCLUSION: Documents and materials presented by Assoc. Prof. Dr. Bogdan Ranguelov meet all the requirements of the Law for the Development of the Academic Staff in the Republic of Bulgaria, the Regulations for its implementation and the corresponding rules for the implementation of the law in the IPC - BAS. The candidate submitted a sufficient number of scientific papers, published after receiving the PhD degree and after the competition for the academic position "Associate Professor". The obtained results based on the research activity of Dr. Bogdan Ranguelov are original and have significant scientific contribution to the studied area. They completely fulfilled the relevant requirements of IPC-BAS for occupation of the academic position "Professor" in the field of competition and are far above them. I strongly support the application and also recommend to the members of the Scientific Jury and to the Scientific Council of the Institute of Physical Chemistry "R. Kaischew" to award to the Assoc. Prof. Dr. Bogdan Stavrev Ranguelov the academic position "Professor" under the direction 4.2. Chemical Sciences "Physical Chemistry".

21/08/2020
Sofia

Reviewer:

(Prof. Dr. Z. Cherkesova-Zheleva,
Member of the Scientific Jury)